

## Dispersion relation for surface plasmons on randomly rough surfaces: A quantum-mechanical approach

D. G. Hall and A. J. Braundmeier, Jr.

*Physics Department, Southern Illinois University, Edwardsville, Illinois 62026*

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We present a quantum-mechanical calculation of the surface plasmon dispersion relation appropriate for randomly rough surfaces. A single-particle Green's function is defined for the surface plasmon propagating on a rough surface and related to its smooth surface counterpart through Dyson's equation. By constructing an approximate self-energy, the plasmon dispersion relation is extracted from the spectral density function.

### I. INTRODUCTION

In recent years there has been considerable theoretical and experimental activity directed towards gaining an understanding of optical effects associated with rough surfaces. Theoretical work has concentrated principally upon calculating the reflectivity of rough surfaces<sup>1-9</sup> and the dispersion relation of surface plasmons (or polaritons) on rough surfaces.<sup>10-12</sup> Discussions of the former have appeared elsewhere and it is the latter which will concern us in this paper. In the last two years, three (classical) calculations of the surface-plasmon dispersion relation on rough surfaces have appeared in the literature. In 1976, Kröger and Kretschmann<sup>10</sup> presented a calculation of the surface-plasmon (SP) dispersion based upon an integral-equation formalism developed by the same authors<sup>1</sup> for the rough-surface reflectivity problem in 1970. Maradudin and Zierau,<sup>11</sup> also in 1976, published a calculation which used a classical Green's-function technique to determine the dispersion relation  $E(k)$ . Both of the above approaches described the surface stochastically, in terms of average quantities and two-point correlation functions. In 1977, Toigo, Marvin, Celli, and Hill<sup>12</sup> reported the details of a calculation of the SP dispersion relation which utilized the Rayleigh method and the extinction theorem. Their resulting equation was deterministic in nature in that  $E(k)$  is obtained for a specific choice of a function which describes the rough sample surface.

Experimental work on randomly rough surfaces is complicated by the difficulties associated with surface characterization. The parameter  $\delta$ , defined to be the root-mean-square height of the surface measured with respect to the average surface, is usually measured (when it is measured) by fringes-of-equal-chromatic-order (FECO) interferometry.<sup>13,14</sup> This technique has been demonstrated to be quite reliable<sup>15</sup> for  $\delta \geq 8 \text{ \AA}$ , but little is known about the reliability of the technique for smaller values of  $\delta$ . Cunningham and Braundmeier<sup>16</sup>

have demonstrated that the difference in near-normal reflectance at  $\lambda = 3500 \text{ \AA}$  between a smooth and a rough silver surface is directly proportional to the value of  $\delta$  (measured by the FECO technique) for  $10 \leq \delta \leq 35 \text{ \AA}$ . The parameter  $\sigma$ , the transverse correlation length, is to some extent a measure of the average separation between similar features on the surface. A precise assignment of a numerical value to  $\sigma$  is somewhat ambiguous and is dependent upon the surface autocorrelation function for a given surface. Bennett has recently reported the results of measurements of the surface autocorrelation function by scanning FECO interferometry.<sup>17</sup> Her measurements indicate that the usual assumption of a Gaussian autocorrelation function is not a very good one for the surfaces she examined. Rasigni *et al.*,<sup>18</sup> have also measured the autocorrelation function (by a different technique) and found it to be more accurately described by a Lorentzian than a Gaussian for their surfaces.

The surface-plasmon dispersion relation has been measured on both randomly rough and periodic surfaces, but measurements made on the former with independently determined surface properties have yet to be reported. Measurements by Braundmeier and Arakawa<sup>19</sup> and subsequent workers<sup>20,21</sup> have shown that the surface plasmon dispersion for  $\text{CaF}_2$  roughened silver films is shifted from the smooth surface values to higher wave numbers (at constant energy) with increasing surface roughness. Similar results have also been obtained for periodically rough surfaces<sup>22-25</sup> and metal gratings.<sup>26-28</sup> The recent theoretical work of Kröger and Kretschmann<sup>10</sup> has been compared by Pockrand and Raether<sup>25</sup> to the SP dispersion relation measured on a surface with a known sinusoidal profile. They found theory and experiment to be in reasonably good agreement for this type of surface.

The calculations of the dispersion relation  $E(k)$  which have appeared in the literature thus far<sup>10-12</sup> have been classical in nature. In this paper we determine  $E(k)$  within a quantum-mechanical frame-

work. The average Green's function is obtained by replacing the proper self-energy in Dyson's equation by an approximate average self-energy. The spectral density for the Green's function is then determined, and  $E(k)$  obtained by examining the structure in the spectral density. Numerical results, while not included in this paper, will be presented in a subsequent paper along with a comparison with other calculations and experimental data which have not yet appeared in the literature.

## II. HAMILTONIAN FOR SURFACE PLASMONS ON A ROUGH SURFACE

The problem we consider is that of a surface plasmon at the interface between a metal and vacuum (both of semi-infinite extent). The metal is assumed to be describable by the dielectric function  $\epsilon(\omega) = 1 - \omega_p^2/\omega^2$  appropriate for the undamped free-electron gas, and to be located in the half space  $z > 0$ . We follow Crowell and Ritchie,<sup>29</sup> Elson and Ritchie,<sup>2</sup> and Celli, Marvin, and Toigo<sup>7</sup> and write the surface plasmon Hamiltonian  $H$  as the sum of two terms

$$H = H_0 + H_1. \quad (1)$$

In the above equation,  $H_0$  is given by

$$H_0 = \frac{1}{8\pi c^2} \int d^3r [\dot{\vec{A}}^2 + \omega_p^2 \Theta(z) \vec{A}^2 + c^2 (\vec{\nabla} \times \vec{A})^2], \quad (2)$$

where  $\vec{A}$  is the vector potential describing surface plasmons on a smooth interface,  $\omega_p$  is the plasma frequency, and  $\Theta(z)$  is the unit step function. Taken by itself,  $H_0$  is the Hamiltonian appropriate for surface plasmons on a planar surface.  $H_1$  is the perturbation term and is given by

$$H_1 = \frac{1}{8\pi c^2} \int d^3r [\Theta(z - \zeta(\vec{R})) - \Theta(z)] \omega_p^2 \vec{A}^2, \quad (3)$$

where  $\zeta(\vec{R})$  gives the height of the surface at position  $\vec{R}$  and  $\vec{R}$  is a two-dimensional coordinate vector. In both of the above,

$$\begin{aligned} \vec{A}(\vec{r}) = \sum_{\vec{k}} \left( \frac{4\pi\hbar c}{L^2 P_k} \right)^{1/2} & \left[ \left( i\hat{k} - \frac{\hat{z}k}{\gamma} \right) e^{-\gamma z} \Theta(z) \right. \\ & \left. + \left( i\hat{k} + \frac{\hat{z}k}{\beta} \right) e^{\beta z} \Theta(-z) \right] \\ & \times e^{i\vec{k} \cdot \vec{R}} (b_{\vec{k}} + b_{-\vec{k}}^\dagger) \end{aligned} \quad (4)$$

with the caret designating a unit vector,  $\vec{k}$  the plasmon wave vector,  $P_k$  a normalization constant, and

$$\gamma^2 = k^2 - \epsilon(\omega)(\omega/c)^2, \quad (5)$$

$$\beta^2 = k^2 - (\omega/c)^2. \quad (6)$$

The creation and annihilation operators,  $b_{\vec{k}}^\dagger$  and  $b_{\vec{k}}$ , satisfy the usual boson commutation relation

$$[b_{\vec{k}}, b_{\vec{k}'}^\dagger] = \delta_{\vec{k}, \vec{k}'}, \quad (7)$$

and the surface plasmon frequency  $\omega$  is related to the wave number  $k$  (for a smooth surface) by

$$k = (\omega/c) [\epsilon(\omega)/(\epsilon(\omega) + 1)]^{1/2}. \quad (8)$$

The integrations necessary in Eq. (2) are easily carried out leaving  $H_0$  in the form

$$\begin{aligned} H_0 = \sum_{\vec{k}} [m(k) + \omega^2 n(k)] + 2 \sum_{\vec{k}} [m(k) + \omega^2 n(k)] b_{\vec{k}}^\dagger b_{-\vec{k}} \\ + \sum_{\vec{k}} [m(k) - \omega^2 n(k)] (b_{\vec{k}} b_{-\vec{k}} + b_{\vec{k}}^\dagger b_{-\vec{k}}^\dagger), \end{aligned} \quad (9)$$

where

$$n(k) = \frac{\hbar}{4cP_k} \left[ \frac{1}{\gamma} \left( 1 + \frac{k^2}{\gamma^2} \right) + \frac{1}{\beta} \left( 1 + \frac{k^2}{\beta^2} \right) \right], \quad (10)$$

$$m(k) = \frac{\hbar}{4cP_k} \left[ \frac{\omega_p^2}{\gamma} \left( 1 + \frac{k^2}{\gamma^2} \right) - \frac{\epsilon\omega^2}{\gamma} \left( 1 - \frac{k^2}{\gamma^2} \right) - \frac{\omega^2}{\beta} \left( 1 - \frac{k^2}{\beta^2} \right) \right], \quad (11)$$

and  $\omega$  is related to  $k$  as in Eq. (8). The form of  $H_0$  is reminiscent of that encountered by Bogoliubov in studying excitations in liquid helium.<sup>30</sup> In that problem, Bogoliubov diagonalized the Hamiltonian by eliminating the scattering term (third term) by means of a rather elegant canonical transformation. The problem is somewhat simpler here because the quantities  $m(k)$  and  $n(k)$  are related such that

$$m(k) - \omega^2 n(k) = 0 \quad (12)$$

when  $\omega$  and  $k$  are related as in Eq. (8), leaving

$$H_0 = \sum_{\vec{k}} [m(k) + \omega^2 n(k)] + 2 \sum_{\vec{k}} [m(k) + \omega^2 n(k)] b_{\vec{k}}^\dagger b_{-\vec{k}}. \quad (13)$$

The normalization constant  $P_k$  is determined by requiring that the second term in  $H_0$  be in standard form  $\sum_{\vec{k}} \hbar\omega_{\vec{k}} b_{\vec{k}}^\dagger b_{-\vec{k}}$ . This procedure yields  $P_k = [(\epsilon^2 - 1)^2 / [\epsilon^2(-\epsilon - 1)^{1/2}]]$ . The final form of  $H_0$ , then, is

$$H_0 = \text{const} + \sum_{\vec{k}} \hbar\omega_{\vec{k}} b_{\vec{k}}^\dagger b_{-\vec{k}}. \quad (14)$$

As we are interested in excitation energies, the constant will be neglected in the work which follows.

The perturbation term  $H_1$  given in Eq. (3) can be reduced in complexity by expanding the quantity in square brackets and retaining only the term linear in  $\zeta(\vec{R})$  as given below.

$$\Theta(z - \zeta(\vec{R})) - \Theta(z) = -\zeta(\vec{R}) \delta(z). \quad (15)$$

Equation (15) has been used recently by Celli *et al.*<sup>7</sup> With Eq. (15),  $H_1$  becomes

$$H_1 = -\frac{1}{8\pi c^2} \int d^3r \zeta(\vec{R}) \delta(z) \omega_p^2 \vec{A}^2. \quad (16)$$

While it appears that the  $z$  integration in Eq. (16) is easily performed, in actuality there is a complication which arises in the form of a discontinuity in the integrand at  $z = 0$ . This point has been addressed directly by Celli *et al.*,<sup>7</sup> who stated that matrix elements of  $H_1$  are then well defined only for states with  $\vec{k} = 0$  (at  $\vec{k} = 0$  the discontinuity disappears, see Ref. 7). Maradudin and Mills,<sup>6</sup> on the other hand, bypassed the difficulty by invoking the evenness of the  $\delta$  function. To be specific, if  $f(z)$  and  $g(z)$  are discontinuous at  $z = 0$ , Maradudin and Mills set forth the rule that

$$\int_{-\infty}^{\infty} f(z) \delta(z) g(z) dz = \frac{1}{2} [f(0^+)g(0^+) + f(0^-)g(0^-)]. \quad (17)$$

Later, Mills<sup>31</sup> found that to achieve agreement between his own calculation of the cross section for the scattering of  $p$ -polarized incident light into  $p$ -polarized scattered light and those of Kröger and Kretschmann<sup>1</sup> and Marvin, Toigo, and Celli,<sup>32</sup> he needed to evaluate the integral according to

$$\int_{-\infty}^{\infty} f(z) \delta(z) g(z) dz = f(0^+)g(0^-). \quad (18)$$

Maradudin and Zierau<sup>11</sup> used the result shown in Eq. (18) in their calculation of  $E(k)$ , but pointed out that there has been no derivation of this result. Because of the lack of justification for Eq. (18), we have chosen to follow a more standard route. It should be recognized, however, that there does appear to be a need for further work in order to understand the questions raised by Eqs. (17) and (18). Our result will suggest that Eq. (17) is more appropriate, but should Eq. (18) turn out to be the correct choice, the results obtained in Sec. III would require only slight modification. Our approach is to replace  $\delta(z)$  in Eq. (16) by

$$\delta(z) = \lim_{l \rightarrow 0} \frac{1}{l\sqrt{\pi}} \exp\left(-\frac{z^2}{l^2}\right). \quad (19)$$

The  $z$  integration is then carried out directly, followed by taking the limit  $l \rightarrow 0$ . Upon performing the remaining integral over  $\vec{R}$ , we obtain

$$H_1 = -\frac{\hbar\omega_p^2}{4cL^2} \sum_{\vec{k}} \sum_{k'} \left(\frac{1}{P_k P_{k'}}\right)^{1/2} \times \left[ \left(\frac{kk'}{\gamma\gamma'} + \frac{k k'}{\beta\beta'} - 2\hat{k} \cdot \hat{k}'\right) \zeta_{\vec{k}, \vec{k}'} \right. \\ \left. \times (b_{\vec{k}} + b_{-\vec{k}}^\dagger)(b_{\vec{k}'} + b_{-\vec{k}'}^\dagger) \right]. \quad (20)$$

In Eq. (20) primed quantities indicate that  $k$  has

been replaced by  $k'$ , and

$$\zeta_{\vec{k}, \vec{k}'} = \int d\vec{R} e^{i(\vec{k} + \vec{k}') \cdot \vec{R}} \zeta(\vec{R}). \quad (21)$$

The sample surface is assumed to be of area  $L^2$  and the limit  $L \rightarrow \infty$  is taken at the end of the calculation (see Ref. 2).

### III. DISPERSION RELATION: GREEN'S-FUNCTION TECHNIQUE

In this section we extract the surface-plasmon dispersion relation from the Hamiltonian formed by using Eqs. (14) and (20) in Eq. (1). This is accomplished by defining a momentum-space single-plasmon Green's function for each of  $H_0$  and  $H$ , and constructing an approximate self-energy  $\Sigma(\vec{k}, E)$ . The two Green's functions are then related through Dyson's equation and the poles of the imaginary part of the Green's function examined to yield  $E(k)$ .

The momentum-space matrix elements of  $(E - H_0 + i\eta)^{-1}$  and  $(E - H + i\eta)^{-1}$  give the Green's functions to be

$$G_0(\vec{k}, E) = \langle 0 | b_{\vec{k}}(E - H_0 + i\eta)^{-1} b_{\vec{k}}^\dagger | 0 \rangle, \quad (22)$$

and

$$G(\vec{k}, E) = \langle 0 | b_{\vec{k}}(E - H + i\eta)^{-1} b_{\vec{k}}^\dagger | 0 \rangle, \quad (23)$$

where  $\eta$  is a positive infinitesimal, and  $|0\rangle$  represents the surface-plasmon vacuum state. By defining the proper self-energy  $\Sigma(\vec{k}, E)$ ,

$$\Sigma(\vec{k}, E) = \langle 0 | b_{\vec{k}} H_1 b_{\vec{k}}^\dagger | 0 \rangle \\ + \sum_{\vec{k}'} \langle 0 | b_{\vec{k}} H_1 b_{\vec{k}'}^\dagger | 0 \rangle G_0(\vec{k}', E) \langle 0 | b_{\vec{k}'} H_1 b_{\vec{k}}^\dagger | 0 \rangle + \dots, \quad (24)$$

in the usual way, we find that  $G$  and  $G_0$  are related by (Dyson's equation)

$$G(\vec{k}, E) = G_0(\vec{k}, E) [1 - \Sigma(\vec{k}, E) G_0(\vec{k}, E)]^{-1}. \quad (25)$$

This can be simplified by noting that Eq. (22) can be written in the form

$$G_0(\vec{k}, E) = (E - \hbar\omega + i\eta)^{-1}, \quad (26)$$

where  $\omega$  is again related to  $k$  through Eq. (8). The expression for  $G$  then becomes

$$G(\vec{k}, E) = [E - \hbar\omega - \Sigma(\vec{k}, E) + i\eta]^{-1}. \quad (27)$$

In order to extract  $E(k)$  from Eq. (27), it is necessary to introduce an approximation to terminate the infinite series for  $\Sigma(\vec{k}, E)$  in Eq. (24). The first term in Eq. (24) is proportional to the height of the average surface. If the average height is taken to be zero, the first term can be neglected. Rewrit-

ing  $H_1$  as

$$H_1 = -\frac{\pi\hbar\omega_p}{2L^2\lambda_p} \times \sum_{\vec{k}} \sum_{\vec{k}'} \left(\frac{1}{P_k P_{k'}}\right)^{1/2} \left[ \left( \frac{kk'}{\gamma\gamma'} + \frac{kk'}{\beta\beta'} - 2\hat{k} \cdot \hat{k}' \right) \zeta_{\vec{k}+\vec{k}'} \times (b_{\vec{k}} + b_{-\vec{k}}^\dagger)(b_{\vec{k}'} + b_{-\vec{k}'}^\dagger) \right], \quad (28)$$

and recognizing that  $\zeta_{\vec{k}+\vec{k}'}$  is proportional to  $\delta$ , the root-mean-square height of the surface, we see that each matrix element of  $H_1$  is proportional to the ratio  $\delta/\lambda_p$ , where  $\omega_p/c = 2\pi/\lambda_p$ .  $\delta$  typically takes on values of 10–50 Å while  $\lambda_p$  has a value of approximately 1000 Å for free-electron metals (for aluminum,  $\hbar\omega_p \approx 15.3$  eV so  $\lambda_p \approx 800$  Å). The ratio  $\delta/\lambda_p$  is thus a small parameter suggesting that  $\Sigma(\vec{k}, E)$  can be well approximated by retaining only the second term in Eq. (24), a term proportional to  $(\delta/\lambda_p)^2$ . The resulting approximate self-energy is

$$\Sigma(\vec{k}, E) = \left(\frac{\pi\hbar\omega_p}{L^2\lambda_p}\right)^2 \times \sum_{\vec{k}' \neq \vec{k}} \frac{|(kk'/\gamma\gamma' + kk'/\beta\beta' - 2\hat{k} \cdot \hat{k}')\zeta_{\vec{k}+\vec{k}'}|^2}{P_k P_{k'} (E - \hbar\omega + i\eta)}. \quad (29)$$

with  $P$  denoting the Cauchy principal value.

The Green's function in Eq. (27), when combined with the approximate self-energy in Eq. (32), provides all of the information necessary to determine  $E(k)$ , the dispersion relation. To carry this out more explicitly, we introduce the spectral density  $\alpha(\vec{k}, E)$  defined by

$$\alpha(\vec{k}, E) = -(1/\pi) \text{Im}[G(\vec{k}, E)]. \quad (33)$$

If the spectral density, for constant  $E$ , is examined as a function of  $k$ , peaks in  $\alpha(k, E)$  give the dispersion relation. For example, if  $\delta=0$ ,  $\langle\Sigma\rangle=0$  and we find

$$\alpha(\vec{k}, E) = (1/\pi) \{ \eta / [(E - \hbar\omega)^2 + \eta^2] \}. \quad (34)$$

Taking the limit  $\eta \rightarrow 0$  gives

$$\alpha(\vec{k}, E) = \delta_D(E(\vec{k}) - \hbar\omega(\vec{k})), \quad (35)$$

To describe the surface in terms of its average properties, we introduce yet another approximation. Instead of substituting Eq. (29) into Eq. (27), we replace  $\Sigma(\vec{k}, E)$  by its ensemble average  $\langle\Sigma(\vec{k}, E)\rangle$ . With Eq. (21) defining  $\zeta_{\vec{k}+\vec{k}'}$ , we find that the average of the absolute square of the Fourier transform of  $\zeta(\vec{R})$  is related to the Fourier transform  $g(\vec{k}' - \vec{k})$  of the surface autocorrelation function according to

$$\langle |\zeta(\vec{k}' - \vec{k})|^2 \rangle = \delta^2 L^2 g(\vec{k}' - \vec{k}). \quad (30)$$

With Eq. (30) the average approximate self-energy becomes

$$\langle\Sigma(\vec{k}, E)\rangle = \left(\frac{\pi\hbar\omega_p}{L}\right)^2 \frac{\delta^2}{\lambda_p^2} \times \sum_{\vec{k}' \neq \vec{k}} \frac{(kk'/\gamma\gamma' + kk'/\beta\beta' - 2\hat{k} \cdot \hat{k}')^2 g(\vec{k}' - \vec{k})}{P_k P_{k'} (E - \hbar\omega + i\eta)}. \quad (31)$$

The limit  $L \rightarrow \infty$  can be taken in Eq. (31) by converting to an integral,  $\sum_{\vec{k}'} \rightarrow (L/2\pi)^2 \int d\vec{k}'$ , however, the sum over  $\vec{k}'$  excludes  $\vec{k}' = \vec{k}$ . Equation (31), for a macroscopic system, must be written as

$$\langle\Sigma(\vec{k}, E)\rangle = \left(\frac{\pi\hbar\omega_p}{2}\right)^2 \left(\frac{\delta}{\lambda_p}\right)^2 P \int d\vec{k}' \left( \frac{(kk'/\gamma\gamma' + kk'/\beta\beta' - 2\hat{k} \cdot \hat{k}')^2 g(\vec{k}' - \vec{k})}{P_k P_{k'} [E(k) - \hbar\omega(k') + i\eta]} \right), \quad (32)$$

or  $E(\vec{k}) = \hbar\omega(\vec{k})$ , as it should. Since  $\langle\Sigma\rangle$  is real for the free-electron gas we are considering, the more general result for the spectral density is

$$\alpha(\vec{k}, E) = \delta_D(E(\vec{k}) - \hbar\omega(\vec{k}) - \langle\Sigma(\vec{k}, E)\rangle). \quad (36)$$

In Eqs. (35) and (36), the subscript  $D$  has been affixed to the Dirac delta function to avoid confusion with the roughness parameter  $\delta$ .

#### IV. DISCUSSION

As mentioned at the end of Sec. I of this paper, a numerical study of Eq. (36) will be deferred until a subsequent paper. For the sake of illustration, if one were to choose a Gaussian autocorrelation function, Eq. (32), the approximate average self-energy, would be written as

$$\langle\Sigma(\vec{k}, E)\rangle = \pi^3 \left(\frac{1}{4}\hbar\omega_p\right)^2 \left(\frac{\delta}{\lambda_p}\right)^2 P \int d\vec{k}' \left( \frac{(kk'/\gamma\gamma' + kk'/\beta\beta' - 2\hat{k} \cdot \hat{k}')^2 \sigma^2 \exp[-\frac{1}{4}(k' - k)^2 \sigma^2]}{P_k P_{k'} [E(\vec{k}) - \hbar\omega(k') + i\eta]} \right). \quad (37)$$

With this "standard guess" for the Fourier transform of the surface autocorrelation function, one must then numerically determine values  $E(\vec{k})$  such that

$$E(k) = \hbar\omega(k) + \langle \Sigma(\vec{k}, E) \rangle. \quad (38)$$

A complete comparison with experiment would require knowledge of  $\delta$ ,  $\sigma$ , and  $g(\vec{k}' - \vec{k})$  for the experimental sample, which is not yet possible. The best one could do would be to treat  $\sigma$  as a fitting parameter within a model (guess) for  $g(\vec{k}' - \vec{k})$  (see, for example, Refs. 31 and 32).

An evaluation of Eqs. (36) or (38) is complicated slightly by the presence of the energy  $E(k)$  inside the sum over  $\vec{k}'$ . This comes about because the Green's-function formalism is based upon infinite order perturbation theory. If we had used Rayleigh-Schrödinger perturbation theory (to second order),  $E(k)$  in the denominator of the right-hand side of Eq. (29) and subsequent equations would have been replaced by  $\omega(k)$ .  $E(k)$  would then be written as

$$E(k) = \hbar\omega(k) + \langle \Sigma(\vec{k}, \omega) \rangle. \quad (39)$$

With regard to the arguments raised in Sec. II of this paper regarding the discontinuities in Eqs. (16)–(18), it was mentioned earlier that the results of Sec. III would be only slightly changed should Eq. (18) emerge as the proper choice. Our procedure of replacing the delta-function in Eq. (16) with a Gaussian, performing the prescribed integration, and then taking the limit of zero width for the Gaussian leads to the same result as that used by Maradudin and Mills<sup>6</sup> [Eq. (17)]. Had we chosen to use the prescription of Eq. (18), our

final result, Eq. (32), would be modified to give the following:

$$\begin{aligned} \langle \Sigma(\vec{k}, E) \rangle = & (\pi \hbar \omega_p)^2 \left( \frac{\delta}{\lambda_p} \right)^2 P \\ & \times \int d\vec{k}' \left( \frac{\hat{k} \cdot \hat{k}' + k k' / \gamma \beta'^2 g(\vec{k}' - \vec{k})}{P_k P_{k'}} [E(k) - \hbar\omega(k') + i\eta] \right). \end{aligned} \quad (40)$$

In our subsequent paper containing numerical results, we shall also examine Eqs. (32) and (40) in an attempt to resolve the questions raised by the discontinuities.

To summarize, we have presented in this paper a calculation of the surface-plasmon dispersion relation appropriate for randomly rough surfaces and based upon a quantum-mechanical formalism. We have extracted the SP  $E(k)$  by defining momentum-space Green's functions for both  $H$  and  $H_0$  and relating them through Dyson's equation. A series of approximations led to an approximate average proper self-energy in terms of which the Green's function could be written in closed form. A numerical study will be published as a subsequent paper in which we will compare the above results with other theoretical approaches as well as new experimental data.

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